Chemical Characteristics of Ice Residual Nuclei

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Ice Nucleation (simple version)

- Complex, since ice can be formed by many processes besides water deposition
- Embryos of ice phase must reach a critical size for nucleation
- Nucleating efficiency varies with temperature (T) and supersaturation

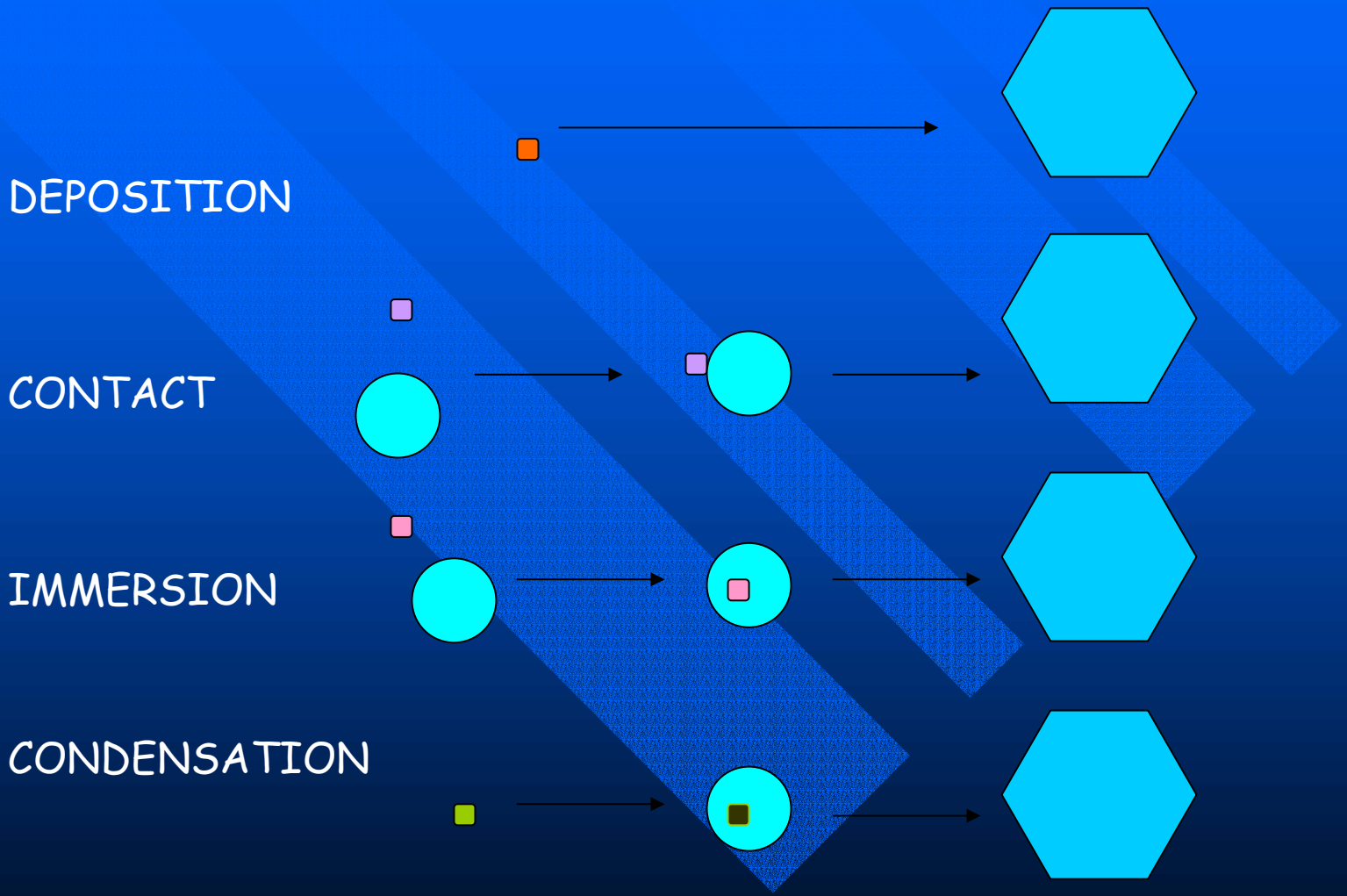
Homogeneous Nucleation

- freezing of pure water drops or liquid aerosol particles
- temperature and RH important
- theory fairly well understood

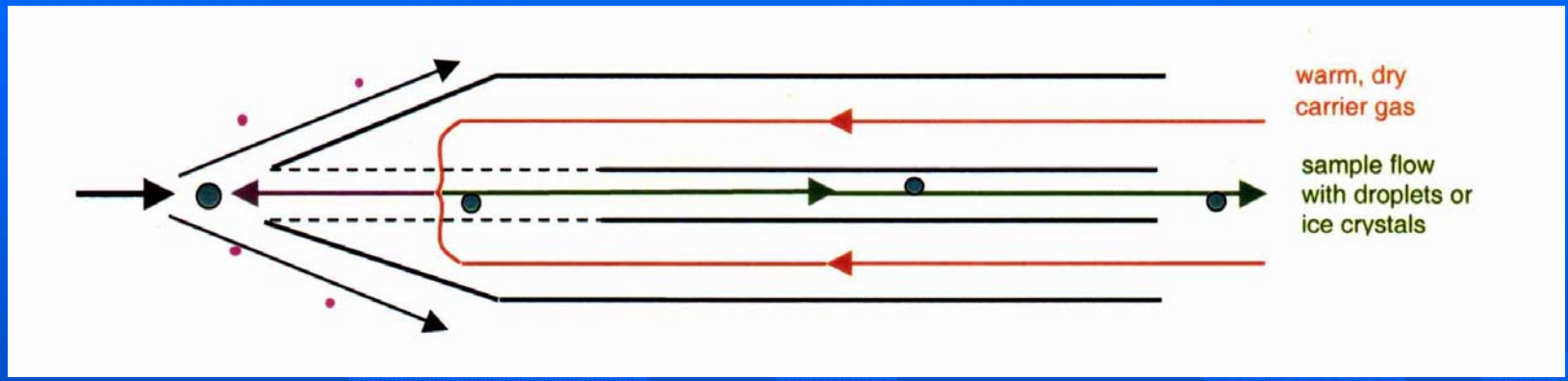
Heterogeneous Nucleation

- ice formation initiated by presence of solid particle
- by freezing (liquid to solid) or deposition (vapor to solid)
- nucleation at specific sites on substrates
- favored nuclei have lattice structure similar to ice
- also temperature and RH sensitive, but can occur at smaller S and warmer T than homogeneous nucleation

Types of Heterogeneous Ice Nucleation



Experiment: CVI on Citation



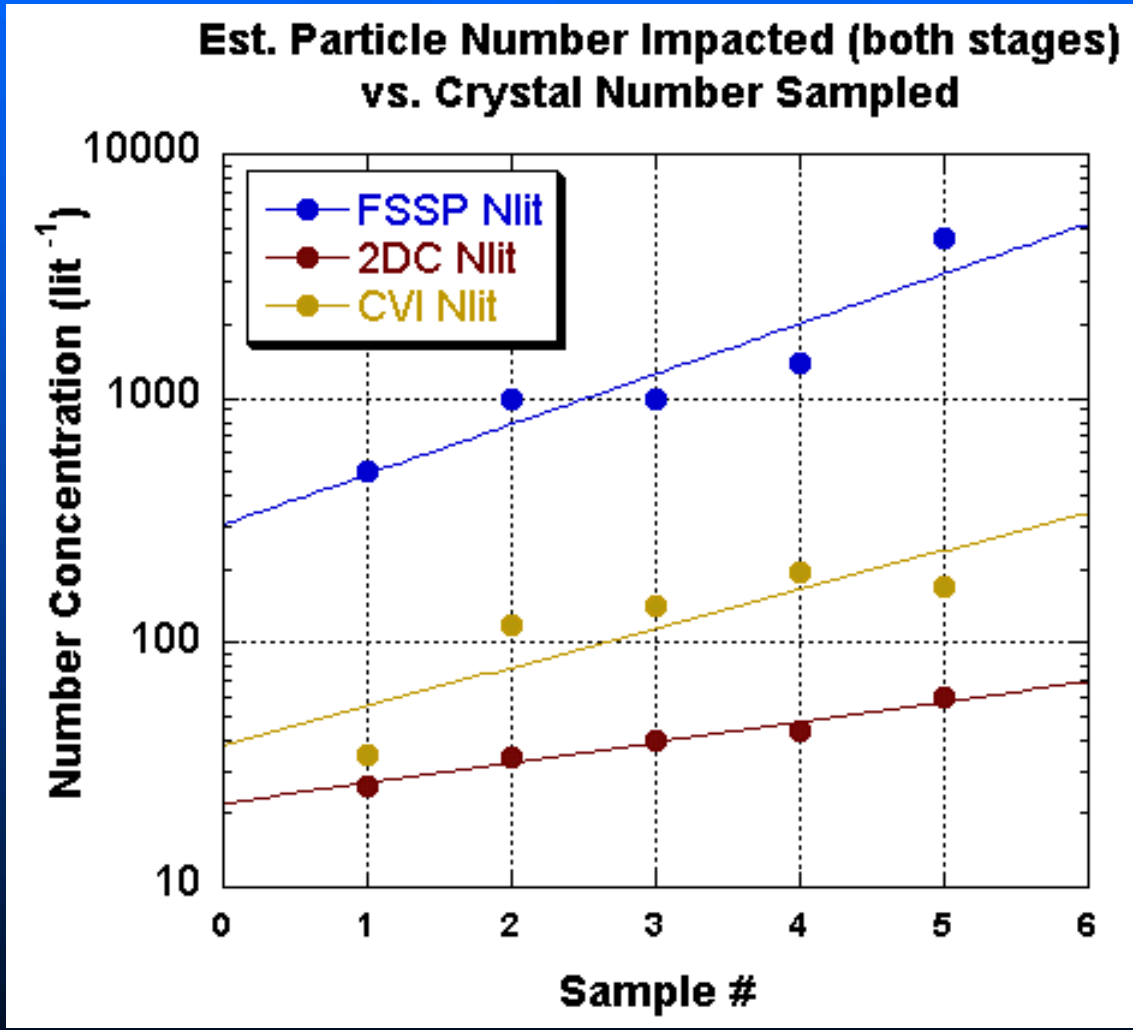
- Crystals were evaporated, their non-volatile residual nuclei & volatile gases measured by various techniques
- Lyman-alpha & TDL (water content crystals > 7 μm)
- CN counter (ice number; only if no large crystals present)
- Ice nuclei (CSU instrument)
- Impactor samples (single particle chemistry via TEM and X-ray analysis)

Particle Classification

(in collaboration with P. Buseck and T. Kojima of ASU)

- **Crustal dust**: silicates (rich in Si, variable Na, Mg, Al, K, Ca, and Fe), carbonates (Mg and/or Ca with large C peak), phosphates
- **Carbonaceous**: ***Soot***: C only. Characteristic morphology (chain aggregates). ***Organic***: amorphous and may contain O, N, and S other than C.
- **Sulfates**: sulfuric acid and ammonium sulfate/bisulfate. S, O only.
- **Metals and Oxides** (Al, Fe, Cr, Ti, Mn, Co, Zn, Cu)
- **Salts**: Chlorides and sulfates of Na, K, Ca and Mg.

Large crystals may scavenge aerosols and break up in CVI; does this affect nuclei N collected on impactor?



CVI number conc. between expected 2D-C conc (min) and FSSP conc (max)

Suggests breakup and/or scavenging doesn't artificially enhance nuclei N in this size range (> 0.07 μm, ρ 1.5)

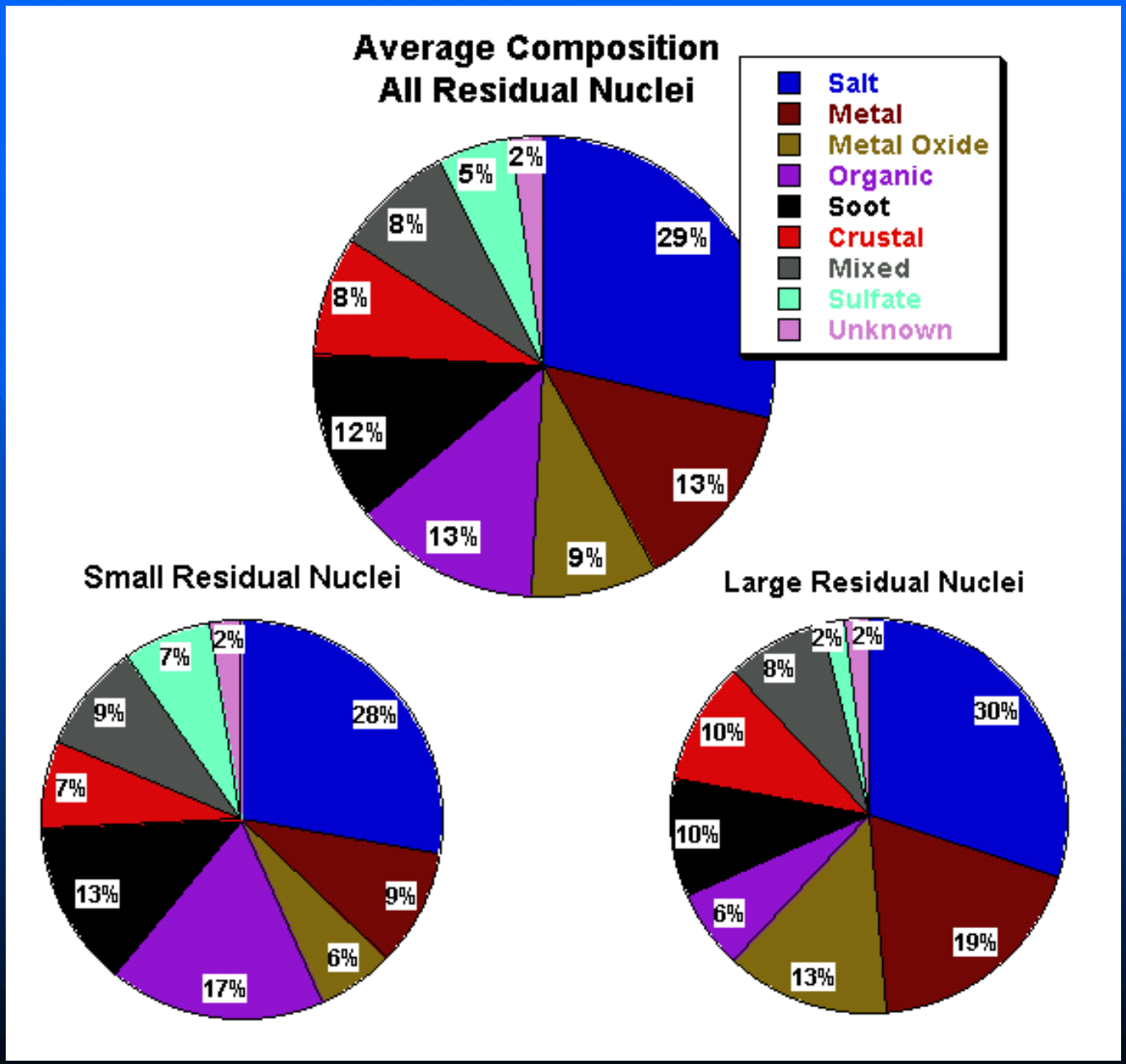
Overall Results

12 samples
6 flights
-57<T<-21

12 small stage
D_{50}=0.07μm

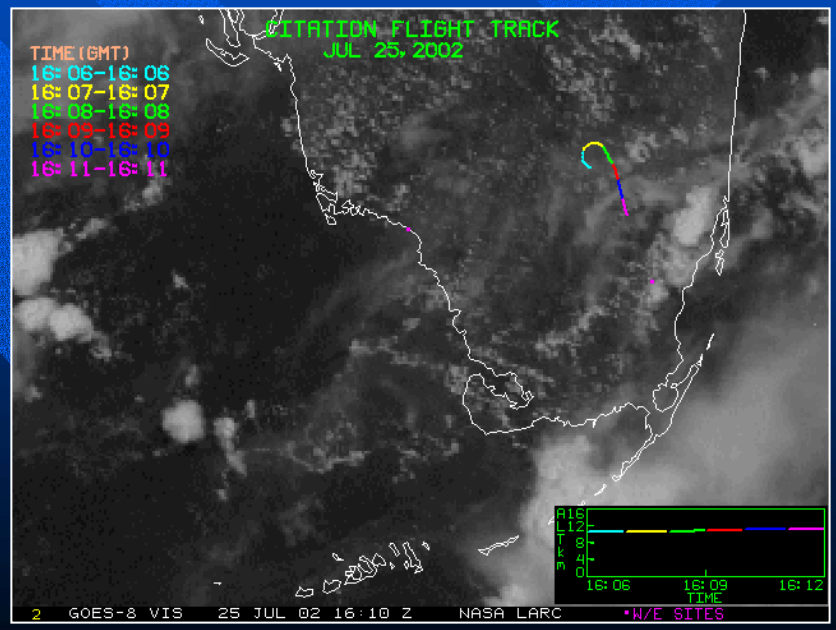
8 large stage
D_{50}=0.40μm

n_{stage}=50
n_{tot}=1000



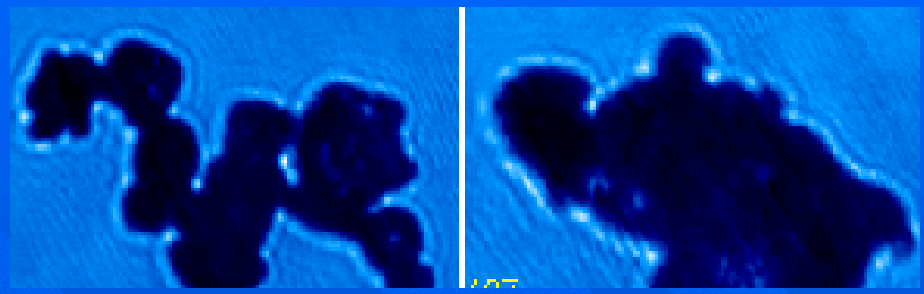
Sample-to-Sample Variability in IN Composition

- Continental vs. Marine, Temperature, Microphysics?
- Case 1: Originating Over Land, Low Altitude
- Case 2: Originating Over Ocean, High Altitude →
- Statistics



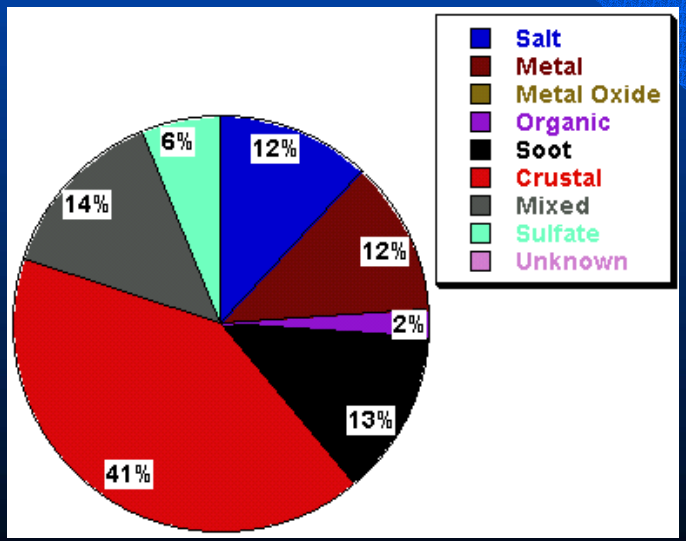
Case 1: Originating Over Land, T -23 C

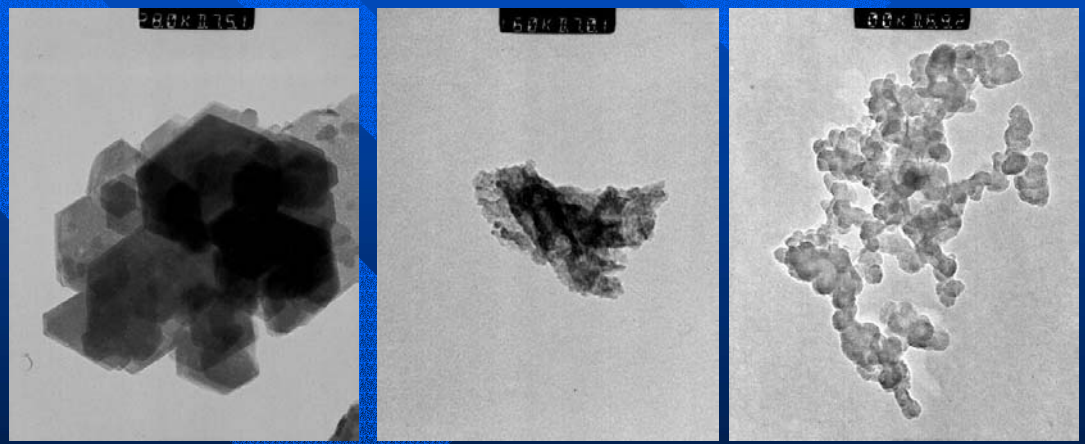
- Continental anvil cirrus has complex cystal habits, aggregates?



CPI Images: Ice Crystals

- Insoluble particles like crustal dust, soot, and metals predominate in these cirrus nuclei

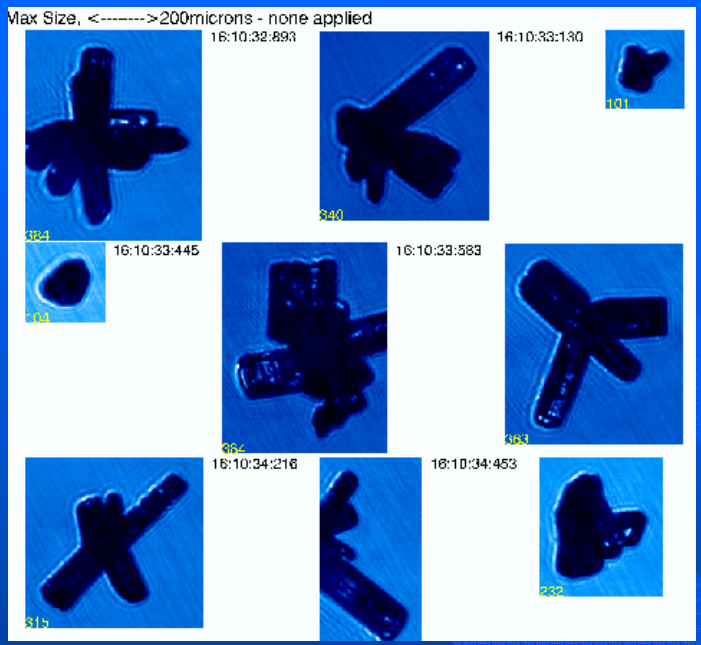




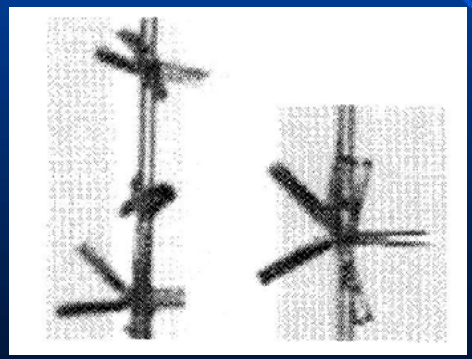
2 µm silicate 0.3 µm metal 0.5 µm soot

Electron Microscope Images: Nuclei

Case 2: Originating Over Ocean, T-42C



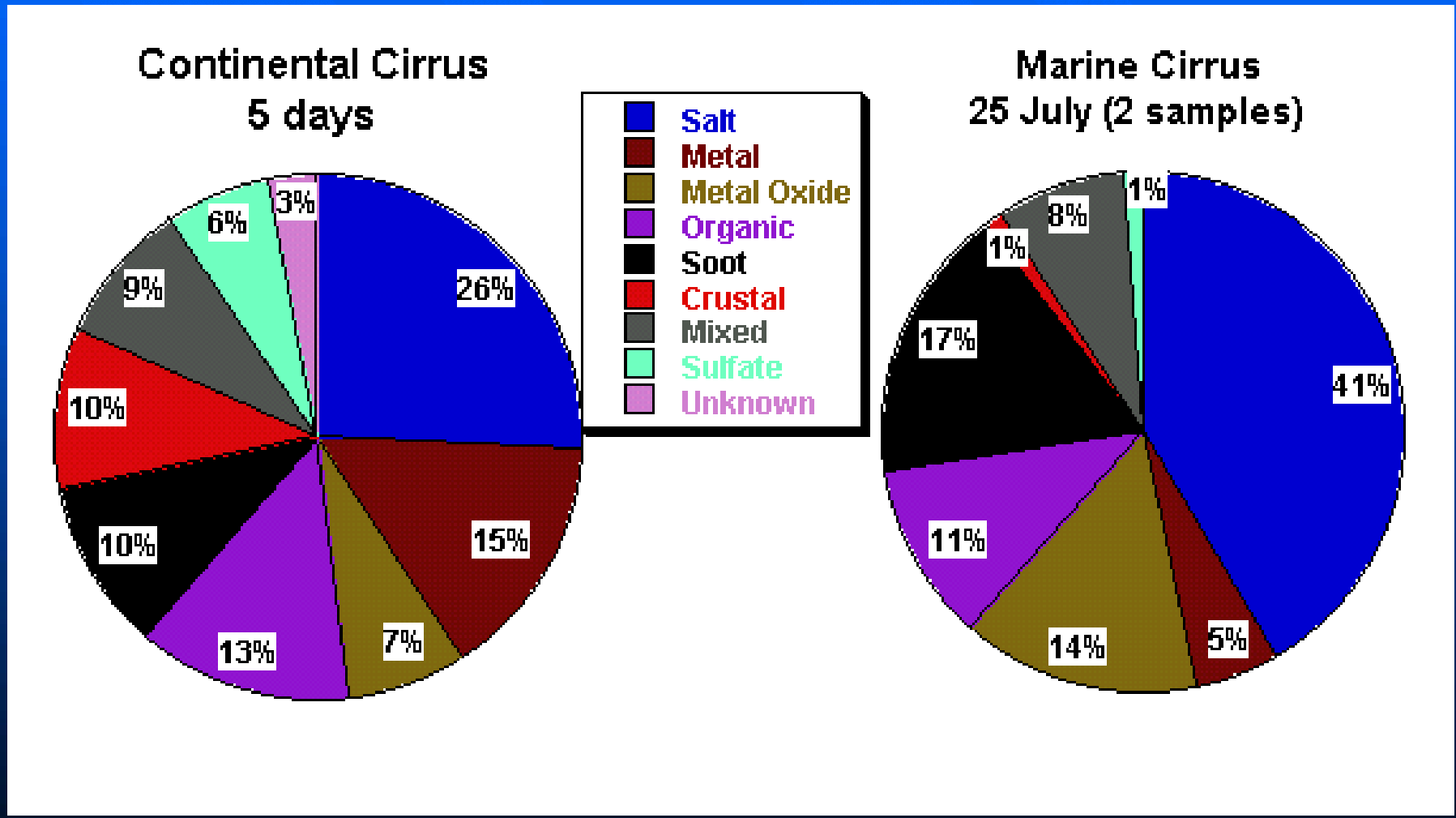
CRYSTAL-FACE bullet rosettes



bullet rosettes grown in Hallett lab

- Maritime anvil cirrus crystals were more pristine in form and almost entirely “bullet rosettes”, as formed by homogeneous freezing in lab (Bailey & Hallett, Q.J.R.M.S., 2002)
- Soluble salts (primarily Na, K, Ca w/ S, Cl) predominated in these cirrus nuclei, virtually no crustal material
- Homogeneous freezing of solution drops may be more prevalent in marine anvils, with fewer heterogeneous ice nuclei

Continental vs. Marine



How is nuclei type related to cirrus parameters?

linear regressions, r> 0.60

	FSSP-N	2DC-N	2DC-MED	PALT	TEMP	IWC
Salt	N 0.18	P 0.14	N 0.57	P 0.68	N 0.70	N 0.20
Metals	P 0.49	P 0.33	P 0.37	N 0.20	P 0.24	P 0.56
Metal Oxide	P 0.28	P 0.49	N 0.01	P 0.14	N 0.12	P 0.27
Organic	P 0.15	P 0.15	P 0.08	P 0.25	N 0.23	P 0.14
Soot	N 0.03	N 0.10	P 0.02	N 0.14	P 0.12	N 0.14
Crustal	N 0.16	N 0.53	P 0.33	N 0.76	P 0.74	N 0.11
Mixed	N 0.12	N 0.20	P 0.33	N 0.47	P 0.46	N 0.18
Sulfate	N 0.24	N 0.19	N 0.24	P 0.17	N 0.17	N 0.21

- Tables for All Nuclei, Large Nuclei, Small Nuclei

Particle Type and Size Relationships

linear regressions, r> 0.60

- At high altitude, low temperature:
  - salts more prevalent for all nuclei sizes-- homogeneous freezing?
- At low altitude, higher temperature:
  - more crustal material (all nuclei sizes)
  - more soot & sulfate (large nuclei sizes)
  - more metals & mixed particles (small nuclei sizes)
- Some weaker correlations with crystal size and IWC

Summary

- Florida anvil cirrus composed of salts, metals, soot and organics; smaller amts of crustal dust, sulfate and mixed particles
- Large nuclei more metals, crustal dust; small nuclei more organics, sulfate
- Salts more prevalent at high altitude, low temperatures; crustal and other nuclei at low altitude, higher temperatures--different nucleation processes?
- Marine cirrus have higher proportion of salt, lack of crustal material, more homogeneous nucleation?

Future Work

- Continue sample analysis to cover more cases and for better statistics
- Analyze ambient samples
- Include crystal habit in the analysis